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PATENT ABSTRACTS OF JAPPAN

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(54) AQUEOUS SOLUTION OR AQUEOUS DISPERSION OF COPOLYMER HAVING WATER-REPELLING PROPERTY, ITS PRODUCTION AND AQUEOUS COATING COMPOSITION CONTAINING THE SAME

PROBLEM TO BE SOLVED: To obtain the subject aqueous solution useful as a coating resin capable of forming a coating film having excellent water-repellence and water resistance by copolymentaing a mixture containing a monomer having a specific structure, a monomer having an alkoxyallyl group, an ethylenic unsaturated monomer, etc.

SOLUTION: The objective aqueous solution is produced by copolymentsing a monomer mixture containing (A) 3-70 wt.% of one or more monomers selected from the monomer of the formula I (R1 is H or methyl; R2 is phenyl or a 1-6C alkyl; n1 is 1-10; n2 is 5-200) and the formula II (R3 is H or methyl; X is H or F; n3 is 1-8; n4 is 1-30), (B) 1-40 wt.% of a monomer having an alkoxysilyl group; (C) 1-25 wt.% of an a, β-ethylenic unsaturated monomer having a functional group selected from carboxyl group, sulfonic acid group and amino group and contains and amino group and (D) 0-95 wt.% of other a, β-ethylenic unsaturated monomers.

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CLAIMS

[Claim(s)]

[Claim 1](a) It is 3 to 70 % of the weight about at least one sort of monomers chosen from a monomer shown by

$$CH^{2} = C - C - (CH^{2}) \frac{H^{2}}{(2!0)} L_{2}$$

$$(1)$$

$$H_{3}$$

$$H_{4}$$

$$H_{5}$$

$$H_{$$

(Among a formula, as for R^1 , R^2 is the same or different in a hydrogen atom or a methyl group, n_1 shows the

integer of 1-10 and n^2 shows the integer of 5-200 for a phenyl group or the alkyl group of the carbon numbers 1-6,

[Formula 2]

 $cH^{z} = c - c - 0 - (cH^{z}) \frac{u_{z}}{c} (cE^{z}) \frac{u_{z}}{c} X$ (II)

(the inside of a formula, and $R^3 - a$ hydrogen atom or a methyl group - X shows a hydrogen atom or a fluorine

are [being (d) and] copolymenzable, and beta- ethylenic unsaturated monomer zero to 95% of the weight. water repellence carrying out copolymerization of the monomeric mixture which contains alpha in which others unsaturated monomer 1 to 25 % of the weight, And the copolymer solution or the water dispersion which has the one sort of functional groups chosen from the group of a sulfonic group and an amino group, and beta- ethylenic The monomer which has alkoxy silyl groups (b) 1 to 40 % of the weight, the (c) carboxyl group, alpha which has stom, n^3 shows the integer of 1-8, and n^4 shows the integer of 1-30, respectively.)

[Claim 2](a) It is 3 to 70 % of the weight about at least one sort of monomers chosen from a monomer shown by

[E slumot], (II) bas (I) slumotarstructural [Formula 3]

respectively.) integer of 1-10 and n^2 shows the integer of 5-200 for a phenyl group or the alkyl group of the carbon numbers 1-6, (Among a formula, as for R^1 , R^2 is the same or different in a hydrogen atom or a methyl group, n^1 shows the

$$CH^{2} = C - C - O - (CH^{2}) - U + X$$
 (II)

[Formula 4]

(the inside of a formula, and R^3 – a hydrogen atom or a methyl group – X shows a hydrogen atom or a fluorine atom, n^3 shows the integer of 1-8, and n_4 shows the integer of 1-30, respectively.)

The monomer which has alkoxy silyl groups (b) 1 to 40 % of the weight, the (c) carboxyl group, alpha which has one sort of functional groups chosen from the group of a sulfonic group and an amino group, and beta-ethylenic unsaturated monomer 1 to 25 % of the weight, And the monomeric mixture which contains 0 to 95 % of the weight for alpha in which others are [being (d) and] copolymerizable, and beta-ethylenic unsaturated monomer, A manufacturing method of the copolymer solution which has the water repellence which adda water and a manufacturing method of the copolymer solution produced by performing a radical polymerization in an organic solvent,

and is characterized by aqueous-ization or forming moisture powder, or a water dispersion.

[Claim 3] A manufacturing method of the copolymer solution according to claim 2 in which an organic solvent

contains alcohol of the carbon numbers 1-8 10% of the weight or more, or a water dispersion. [Claim 4] After mixing resin (B) solution which does not contain a water-repellent group in a copolymer (A) solution and in which aqueous-izing or water decentralization is possible, water and a neutralizer are added, and it is a manufacturing method of aqueous-izing, the copolymer solution according to claim 2 or 3 formed into moisture

powder, or a water dispersion. [Claim 5] a copolymer (A) solution – after [or] adding a surface-active agent which has a water-repellent group in a copolymer (A) and a mixed solution of (B) – a manufacturing method of copolymer solution of claims 2 thrulor 4 addeedus-izing or given in any 1 paragraph which carries out moisture powder, or a water dispersion.

[Claim 6] A manufacturing method of the copolymer solution according to claim 5 whose surface-active agent is

what has a water-repellent group shown by following structural-formula (VI), or a water dispersion.

(R 10 shows a phenyl group or the alkyl group of the carbon numbers 1-6 among a formula, and 7 shows the integer of 5-200, respectively.)

[Claim 7]A manufacturing method of the copolymer solution according to claim 5 whose surface-active agent is what has a water-repellent group shown by following structural-formula (VII), or a water dispersion.

(Y shows a hydrogen atom or a fluorine atom among a formula, and n⁸ shows the integer of 1-30, respectively.) [Claim 8]A manufacturing method of copolymer solution which obtains copolymer solution according to claim 1 or

a water dispersion using an emulsion polymerization method, or a water dispersion. [Claim 9](a) It is 3 to 70 % of the weight about at least one sort of monomers chosen from a monomer shown by

following structural-formula (I) and (II), [Formula 7]

(Among a formula, as for R^1 , R^2 is the same or different in a hydrogen atom or a methyl group, n 1 shows the integer of 5-200 for a phenyl group or the alkyl group of the carbon numbers 1-6, respectively.)

$$c H' = C - C - O - (c H') \frac{u_2}{v} (c F_1) \frac{h^4}{v} X$$
 (II)

(the inside of a formula, and R^3 – a hydrogen atom or a methyl group – X shows a hydrogen atom or a fluorine

atom, n³ shows the integer of 1-8, and n⁴ shows the integer of 1-30, respectively.)

A monomer which has alkoxy silyl groups (b) 1 to 40 % of the weight, the (c) carboxyl group, alpha which has one sort of functional groups chosen from a group of a sulfonic group and an amino group, and beta- ethylenic unsaturated monomer 1 to 25 % of the weight, And a distemper constituent which contains copolymer solution or a water dispersion which has the water repellence which carries out copolymerization of the monomeric mixture which contains alpha in which others are [being (d) and] copolymerizable, and beta- ethylenic unsaturated

monomer zero to 95% of the weight as a vehicle component. [Claim 10]The distemper constituent according to claim 9 in which a monomeric mixture contains a monomer

which has the (e) carbonyl group one to 30% of the weight. [Claim 11]The distemper constituent according to claim 10 which contains a hydrazine derivative which has at least two -NH-NH_{Σ} content groups in one molecule as a cross linking agent.

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the water dispersion which has water repellence.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001] [Field of the Invention] This invention relates to copolymer solution useful as paint resin which can form the coat which was excellent in water repellence and a water resisting property in detail or a water dispersion, a manufacturing method for the same, and the distemper constituent containing this about the copolymer solution or

[0002] [Description of the Prior Art] Although the method of giving water repellence to a dry paint film by mixing in paints the water repellent of low molecular weight which is represented by poly dimethylsiloxane was well known from the former, there was a problem of water repellence falling by temporality. On the other hand, it is possible by taking the method of introducing a water-repellent group into resin like a silicone modification acrylic resin to

obtain the high water repellent coating film of water-repellent durability. [0003] Although aquosity-ization of such water-repellent resin is strongly called for also from on accident prevention from the field of the health at the time of air pollution prevention and paint, generally conventional water-repellent resin has strong hydrophobicity, and aqueous-izing and moisture powder are difficult for it. As a means to solve such a problem, the method of aquosity-izing by carrying out the emulsion polymerization of the alpha and beta- ethylenic unsaturated monomer to an organopolysiloxane macro monomer is indicated, for example by JP, Z-150475, A. however, a surface-active agent with hydrophilic nature high in this method – not using it — until it does not obtain but sufficient water repellence is shown — **** — it did not result. Although there was also a method of carrying out self-emulsification of the water-repellent resin by hydrophilic functional group introduction of a carboxyl group etc., extremely high resin acid value was needed, and it was not able to be said

to be a desirable method from the waterproof field.

[Means for Solving the Problem]As a result of inquiring wholeheartedly that the above-mentioned problem should be solved, by making indispensable a monomer which has a monomer and alkoxy silyl groups which give water repellence and a water repellence, this invention persons found out that aqueous resin which can give water repellence and a water

resisting property excellent in a coat was obtained, and reached this invention. [0005]That is, this invention is 3 to 70 % of the weight about at least one sort of monomers chosen from a

monomer shown by structural-formula (I) and (II) of (a) following, [0006]

[0007](Among a formula, as for R^1 , R^2 is the same or different in a hydrogen atom or a methyl group, n^1 shows the integer of 5-200 for a phenyl group or the alkyl group of the carbon numbers 1-6, respectively.)

$$\begin{array}{c} O \\ \parallel \\ CH^{x} = C - C - O - (CH^{x}) \xrightarrow{u_{3}} (CE^{x}) \xrightarrow{u_{+}} X \qquad (11) \\ I \\ E_{2} \\ [Louinis 10] \end{array}$$

[0100]

[8000]

[0009](the inside of a formula, and R^3 – a hydrogen atom or a methyl group – X shows a hydrogen atom or a fluorine atom, n^3 shows the integer of 1-8, and n^4 shows the integer of 1-30, respectively.) The monomer which has alkoxy silyl groups (b) 1 to 40 % of the weight, the (c) carboxyl group, alpha which has one sort of functional groups chosen from the group of a sulfonic group and an amino group, and beta- ethylenic unsaturated monomer 1 to 25 % of the weight, And the copolymer solution or the water dispersion which others water repellence carrying out copolymerization of the monomer mixture which contains alpha in which others are [being (d) and] copolymerizable, and beta- ethylenic unsaturated monomer zero to 95% of the weight, And the distemper constituent which contains the manufacturing method and this copolymer solution, or a water dispersion as a vehicle component is provided.

[Embodiment of the Invention]At least one sort of monomers (a) chosen from the monomer shown by above structural-formula (I) and (II) in this invention, Water repellence is given to a copolymer and any 1 way of the monomer (a-2) containing the fluoride shown by the monomer (a-1) and the above-mentioned structural-formula (I), or both can be used. [101] which have a polysiloxane chain shown by the above-mentioned structural-formula (I), although R^1 is a hydrogen atom or a methyl group and R^2 is a phenyl group or an alkyl group of the carbon numbers 1-6, there is not necessarily the necessarily that it is mutually the same. n^1 — the integer of 1-10, and n^2 — 5-200 — it is an integer of 15-150 preferably. Since the hydrophilic nature of a copolymer runs short and it becomes poor [water solubility or water dispersibility] when water repellence with n^2 sufficient by less than five is not acquired but it exceeds 200 dispersibility] when water repellence with n^2 sufficient by less than five is not acquired but it exceeds 200

conversely, it is not desirable. As an example of this monomer (a-1), "Silaplane FM-0711", "Silaplane FM-0721", "Silaplane FM-0725" (all are the Chisso Corp. make), etc. are mentioned with a commercial item. [0012]In the monomer (a-2) shown by the above-mentioned structural-formula (II), \mathbb{R}^3 is a hydrogen atom or a methyl group, and X is a bydrogen atom or a structural description of \mathbb{R}^3 is an integer of \mathbb{R}^3 is an integer of \mathbb{R}^3 is an integer of \mathbb{R}^3 in an integer of \mathbb{R}^3 is an integer of \mathbb{R}^3 in an integer of \mathbb{R}^3 in an integer of \mathbb{R}^3 is an integer of \mathbb{R}^3 in \mathbb{R}^3 in an integer of \mathbb{R}^3 in \mathbb{R}^3 in \mathbb{R}^3 in \mathbb{R}^3 integer of \mathbb{R}^3 in \mathbb{R}^3 in

methyl group, and X is a hydrogen atom or a fluorine atom. n³ is an integer of 1-8 and n⁴ is an integer of 1-30. If this monomer and it will become poor [water solubility or water dispersibility], it is not desirable, as the example of this monomer (a-2) – 2, 2, and 2-trifluoroethyl (meta-) acrylate. 2,2,3,3-tetrafluoro propyl (meta) acrylate, 2-(pentadecafluorooctyl) ethyl (meta) acrylate, in commercial items, such as 2-(nonadecafluorodecyl) ethyl (meta) acrylate, in commercial items, such as 2-(nonadecafluorodecyl) ethyl (meta) acrylate, "FAMAC" (made by Nippon Mektron, Ltd.), "screw coat 8FM", "screw coat 17FM" (all are the OSAKA acrylate, "FAMAC" (made by Nippon Mektron, Ltd.), "screw coat 8FM", "screw coat 17FM" (all are the OSAKA

ORGANIC CHEMICAL INDUSTRY, LTD. make), etc. are mention has the alkoxy silyl groups shown by following structural-formula (III), [0014]

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[0015] (Among a formula, as for R^5 and R^6 , R^4 is the same or different in the alkyl group of the carbon numbers 1-6, or the alkoxyl group of the carbon numbers 1-4, respectively.) When n^5 is two or more, R^5 comrade and a comrade's R^6 may be the same, or may differ from each other.

The monomer shown by following structural-formula (IV) and (V) as the example of representation can be

.benotinen

[9100]

[0017](Inside of a formula, and A) [0018]

_____ = xx -o-ɔ-

 $[0019]^{*****}$. R^7 shows a hydrogen atom or a methyl group, and R^8 shows the divalent aliphatic-saturated-hydrocarbon group of the carbon numbers 1-6, respectively. R^4 , R^5 , R^6 , and n^5 have the respectively same meaning as the above.

[0020]

Eomonia 1

$$CH^{2} = CH - (B_{0}) \frac{B_{0}}{|} B_{0}$$

$$(A)$$

$$B_{0}$$

[0021](R^3 shows the divalent aliphatic-saturated-hydrocarbon group of the carbon numbers 1-6 among a formula, and n^6 shows 0 or 1.) R^4 , R^5 , R^6 , and n^5 have the respectively same meaning as the above. In the above-mentioned structural-formula (IV) and (V), as a divalent aliphatic-saturated-hydrocarbon group of the carbon numbers 1-6 shown by R^8 and R^9 , The alkylene group of a straight chain or the letter of branching, for example, methylene, a hexamethylene, propylene, 1,2-, 1,3- or 2,3-butylene, tetramethylene, athly group of the carbon numbers 1-6 shown by R^5 and R^6 , The alkyl group of a straight chain or the letter of branching, for example, methyl, ethyl, isopropyl, isopropyl, n-, i-, sec- or tert-butyl, n-pentyl, isopentyl one, n-hexyl, isohexyl, 1-methylpentyl, etc. are mentioned. n-heptyl, 2-ethylhexyl, n-nonyl, n-decyl, etc. other than what was illustrated as an alkyl are mentioned. n-heptyl, 2-ethylhexyl, n-nonyl, n-decyl, etc. other than what was illustrated as an alkyl

[Formula 15] [0022]It is A among monomers of the above-mentioned structural-formula (IV). [0023] butoxy, n-pentoxy, isopentoxy, n-hexyloxy, isohexyloxy, n-octyloxy, etc. are mentioned. straight chain or letter of branching, for example, methoxy, and ethoxy **n-propoxy, isopropoxy, n-, i-, sec- or tert- R^4 are mentioned further. As an alkoxyl group of the carbon numbers 1-10 shown by R^5 and R^6 , Alkoxyl group of acoup of the carbon numbers 1-6 shown by $\rm R_2$ and $\rm R_6$ as an alkyl group of the carbon numbers 1-10 shown by

acryloyloxypropylmethyldimethoxysilane, gamma-(meta) acryloyloxypropyl methyldiethoxysilane, etc. can acryloyloxypropyl trimethoxysilane, gamma-(meta) acryloyloxypropyl triethoxysilane, gamma-(meta) [0024]Come out and as a certain thing, for example beta-(meth)acryloyloxy ethoxysilane, gamma-(meta)

illustrate surtably.

[0025]It is A among the monomers of the above-mentioned structural-formula (IV). [0026]

[0027]coming out – as a certain thing – for example [0028]

[Formula 17]

[0030]As a monomer of the above-mentioned structural-formula (V), vinyltimetoxysilane, vinyltriethoxysilane, etc. .benotinem zi ****[6200]

2-ethylhexyl acrylate, acrylic acid (meta) n-octyl, (Meta) Decyl acrylate, acrylic acid (meta) lauryl, acrylic acid acrylic acid (meta) n-butyl, (Meta) Acrylic acid i-butyl, acrylic acid (meta) t-butyl, acrylic acid (meta) hexyl, (Meta) example, methyl acrylate (meta), ethyl acrylate (meta), acrylic acid (meta) n-propyl, (Meta) Acrylic acid isopropyl, [0032]In this invention, as other copolymerizable alpha and a beta- ethylenic unsaturated monomer (d), For acrylate, and t, for example, - Butylamino ethyl (meta) acrylate etc. are mentioned. monomer which has an amino group, they are dimethylaminoethyl (meta) acrylate, diethylaminoethyl (meta) acrytoxyethyl sulfonic acid etc. are mentioned as a monomer which has a sulfonic group, for example. As a itaconic acid anhydride, a succinic anhydride, phthalic anhydride, etc., etc. are mentioned, and 2-(meta) anhydrous 2 organic-functions carboxylic acid (for example, a maleic anhydride.) An equimolar addition with acrylate, an unsaturated monomer which has 5-carboxyl pentyl (meta-) acrylate and a hydroxyl group, and crotonic acid, itaconic acid, maleic acid, Fumaric acid, 2-carboxyl ethyl (meta) acrylate, 2-carboxyl propyl (meta) water solubility or water dispersibility of a copolymer, and has a carboxyl group, For example (meta), acrylic acid, and an amino group in this invention, and beta- ethylenic unsaturated monomer (c), As a monomer which raises [0031]alpha which has one sort of functional groups chosen from a group of a carboxyl group, a sulfonic group, are mentioned, for example.

(meta) stearyl, (Meta) Acrylic acid cyclohexyl, acrylic acid (meta) isobomyl, (Meta) Acrylic acid methoxy ethyl, acrylic acid (meta) ethoxybutyl, (Meta) Acrylic acid methoxy butyl, acrylic acid (meta) ethoxybutyl, (Meta) Acrylic acid acid (meta) ethoxypropyl, acrylic acid (meta), Ardroxypropyl, acrylic acid (meta) A-hydroxybutyl, a polyethylene glycol, and a polypropylene glycol, and acrylic acid (meta) A-hydroxybutyl, a polyethylene glycol, and a polypropylene glycol, and acrylic acid (meta), Which (meta) A-hydroxybutyl, a polyethylene glycol, and a polypropylene glycol, and acrylic acid (meta), Which (meta) acrylic ester; Ethyl vinyl ether, n-propylvinyl ether, Icopropylvinyl ether, n-butylvinyl ether, n-butylvinyl ether, hexylvinyl ether, octylvinyl ether, cyclohexylvinyl ether, Vinyl ether, arch as phenylvinyl ether, Jinyl acetate, Vinyl ester, arch as lactic acid vinyl, butanoic acid vinyl, and caproic acid vinyl; Propenyl ester, arch as isopropenyl acetate, and I for the purpose of one sort or two sorts or vinyltoluene, alpha - KURORU styrene etc. are mentioned, and I for the purpose of one sort or two sorts or

more], these can be chosen suitably and can be used.

[0033]A copolymentation ratio of the above-mentioned monomers preferably a monomer (a) three to 70% of the weight, it is [monomer \ (b)] 20 to 70 % of the weight zero to 95% of the weight one to 40 % of the weight, it is [monomer \ (b)] 20 to 70 % of the weight zero to 95% of the weight about 5 to 15 % of the weight in a monomer (c) three to 20% of the weight preferably one to 40% of the weight about 5 to 15 % of the weight, and a monomer (d). Since aqueous-izing of a copolymer or moisture powder-ization will become difficult if water repellence sufficient in less than 3 % of the weight is not acquired but a monomer (a) exceeds 70 % of the weight conversely, it is not desirable. Since it will arise and become easy to gel hydrolysis and a self-condensation reaction of alkoxy silyl groups to a polymerization process of a copolymer, or an aqueous-izing and dispersibility of copolymer aqueous (moisture powder) liquid] in less than 1 % of the weight or water the weight conversely, it is not desirable. Since sufficient water repellence will not be acquired but a water resisting property will also fall remarkably further if aqueous-izing of a copolymer or moisture powder-ization resisting property will also fall remarkably further if aqueous-izing of a copolymer or moisture powder-ization resistion and a monomer (c) exceeds 25 % of the weight conversely in less than 1 % of the weight, it is

not desirable. [0034] Copolymer solution or a water dispersion of this invention can be manufactured using a mixture of above-mentioned monomer (a) - (d) by methods, such as an emulsion polymerization method which use a neutralizer and uses aqueous-izing or a method of forming into moisture powder, and ** surface-active agent after ** solution polymerization. From a water-repellent and waterproof point of a coat obtained [especially] by this invention. ** Add water and a neutralizer to this and provide it with a manufacturing method of aqueous-izing, copolymer solution which carries out moisture powder, or a water dispersion, after performing a method, i.e., an above-mentioned monomeric mixture, in an organic solvent, performing a radical polymerization under polymerization

initiator existence and obtaining a copolymer (A) solution.

[0035]Copolymer solution or a water dispersion of this invention can be manufactured without using a ****** surface-active agent for water repellence or a water resisting property for an adverse effect according to the

manufacturing method of this invention. [0036]In this invention method, as an organic solvent used at the time of a radical polymerization, it is desirable to be able to use an alcohol system, a cellosolve system, a carbinol system, a cellosolve system, a cellosolve system, a cellosolve acetate system, etc., and to contain alcohol of the carbon numbers 1-8 30% of the weight or more especially preferably 10% of the weight or more in an organic solvent. As a radical polymerization initiator, for example 2,2-azobisisobutyronitrile, An initiator of peroxide systems, such as also polymerization initiators, such as 2,2-azobis (2,4-dimethylvaleronitrile), or lauryl peroxide, t-butyl par 2-ethyl hexanate, and benzoyl peroxide, can be used. This radical polymerization start agent peroxide, t-butyl par 2-ethyl hexanate, and benzoyl peroxide, can be used. This radical polymerization start agent

concentration has 0.3 to 10 preferred weight section to monomer 100 weight section. [0037]In this invention method, as a neutralizer used on the occasion of aqueous-izing or moisture powder, When a monomer (c) has a carboxyl group and a sulfonic group, For example, monomethylamine, directhylamine, friethylamine, monoethylamine, disopropylamine, Triisopropylamine, monoethylamine, diethanolamine, Triisopropylamine, monobutyl amine, dibutylamine, tributylamine, Monoethanolamine, diethanolamine,

triethanolamine, Amine, such as dimethylamino ethanol and diethylamino ethanol, When ammonia, sodium hydrate, etc. can be used and a monomer (c) has an amino group, organic acid, such as inorganic acid, such as chloride, sulfuric acid, and phosphoric acid, formic acid, acetic acid, propionic acid, triengly despite acid, social (propionic acid, social, acid, acid,

trimethylacetic acid, acrylic acid (meta), lactic acid, can be used, for example.

[0038]A copolymer (A) obtained by the above-mentioned radical polymentation Aqueous-izing or when moisture powder is carried out, For example, after neutralizing by adding a neutralizer, agitating a copolymer (A) solution, it is also possible to carry out phase conversion of the copolymer (A) which added water or was neutralized by adding gradually underwater, but. In this case, since hydrolysis and a condensation reaction of alkoxy silyl groups advance quickly and there is a possibility of thickening and gelling, in order to prevent it, if is desirable to perform neutralization and water addition as much as possible for a short time. Although time from neutralizer addition to phase conversion by water changes with a reaction vessel, chuming conditions, and ambient temperature, appearably, generally, generally, it is convenient to consider it as less than 10 hours preferably for less than 24 hours. It is specifically, generally, it is convenient to consider it as less than 10 hours preferably for less than 24 hours. It is copolymer (A) solution, and adding a neutralizer after that, without making the above-mentioned copolymer (A) solution, and adding a neutralizer after that, without making the above-mentioned copolymer (A) solution is blended after moisture powder according to this method, and a siland groups, and promotes bridge construction is blended after moisture powder according to this method, and a siland group may promotes bridge construction is blended after moisture powder according to this method, and a siland group may

exist stably, thickening and gelling can be prevented. [0039]In this invention method, after mixing resin (B) solution which does not contain a water-repellent group in a copolymer (A) solution and in which aqueous-izing or water decentralization is possible, water and a neutralizer are added, and into this mixture, it can water-grit, or can moisture-powder-ize, and can manufacture into it. [0040]As this resin (B), it it mixes with a copolymer (A) enough, there will be no restriction in particular, For example, a copolymer obtained by carrying out copolymerization of a monomer which gives water solubility/water dispersibility, and the other monomers, and a copolymer obtained by choosing it as said monomer (c) and a monomer (b), and (d) suitably from listings, and specifically carrying out copolymerization to them can be used. A monomer (b), and (d) suitably from listings, and specifically carrying out copolymerization to them can be used. A

copolymentzation reaction can be performed like a copolymer (A). [0041]As for a using rate of resin (B), when using the above-mentioned resin (B), it is preferably desirable in sum total resin solid content with a copolymer (A) to make it become 80 or less % of the weight 95 or less % of the weight. Since sufficient water repellence cannot be acquired if a using rate of this resin (B) exceeds 95 % of the

(A) solution or a copolymer (A), and resin (B), it can water-grit or water decentralize. [0043]As this surface-active agent, what has a water-repellent group shown, for example by following structural-

formula (VI) or (VII) can be used. [0044]

[9400]

 $[0045](R^{10}$ shows a phenyl group or the alkyl group of the carbon numbers 1-6 among a formula, and n^7 shows the integer of 5-200, respectively.)

[0047](Y shows a hydrogen atom or a fluorine atom among a formula, and n⁸ shows the integer of 1-30, respectively.) Specifically as a surface-active agent shown by following structural-formula (VI), polyether modified silicone oil, alkyl modified silicone oil, etc. are mentioned, for example. Specifically as a surface-active agent shown by following structural-formula (VII), perfluoroalkyl carboxylate, a perfluoro alkyl-sulfonic-acid salt, etc. are mentioned, for example. As for the addition of this surface-active agent, it is desirable that it is 5 or less % of the weight preferably 10 or less % of the weight to resin solid content. Since the water repellence and the water resisting property of a cost which will be obtained if this addition exceeds 10 % of the weight fall, it is not

0048]On the other hand, by this invention, an emulaitier is used for this through a manufacturing method of the aforementioned **, i.e., water, distributed emulaification of the mixture of said monomer (a) - (d) is carried out, and an emulaion polymentation method performed by adding a water-soluble polymentation initiator and heating at 50-90 ** is also provided. If a redox initiator is used, it is also possible to carry out at a room temperature. As an emulaitier, for example Anionic surface active agents, such as sulfate of higher alcohol, and an alkyl-sulfonic-acid salf, Monionic surface active agents, such as various alkyl ether of a polyoxyethylene, alkyl ester, and alkyl allyl ether, a reactive surface active agent which has a polymerization nature unsaturation group, etc. are used. As a ether, a reactive surface active agent which has a polymerization nature unsaturation group, etc. are used. As a polymerization initiator, hydrogen peroxide, ammonium persulfate, cumene hydroperoxide, or a water-soluble

redox initiator is used, for example. [0049]Subsequently, in this invention, a distemper constituent which contains copolymer solution or a water

dispersion manufactured as above-mentioned as a vehicle component is provided. [0050] into a mixture of monomer (a) - (d) used for manufacture of the above-mentioned copolymer solution or a water dispersion, a monomer (e) which has a carbonyl group can be further contained five to 20% of the weight if needed from points, such as a water resisting property. [0051] As a monomer (e) which has a carbonyl group, For example, an acrolein, discetone acrylamide, die

introduced into a copolymer. Hydrazide groups and a semicarbaside group are contained in a -NH-NH $_{
m 2}$ content

group here. [0053]As this hydrazine derivative, for example Oxalic acid dihydrazide, malonic acid dihydrazide, Glutaric acid dihydrazide, Galurated-fat fellows carboxylic acid dihydrazide, such as sebacic acid dihydrazide; Maleic acid dihydrazide, Monoolefin nature unsaturated-dicarboxylic-acid dihydrazide, auch as fumaric acid dihydrazide, Monoolefin nature Phthalic acid, Terephthalic acid or isophthalic acid dihydrazide, and dihydrazide and itaconic acid dihydrazide; Phthalic acid, Terephthalic acid or isophthalic acid dihydrazide, and dihydrazide of pyromellitic acid, TORIHIDORAJIDO or tetrahydrazide; Mit RIROTORI hydrazide, Trihydrazide of pyromellitic acid, TORIHIDORAJIDO, ethylene-diamine-tetrascetic acid tetrahydrazide, 1,4,5,8-naphthoic acid tetrahydrazide, polyhydrazide which makes a low-grade polymer which has a carboxylic acid lower-alkyl-eater group come to react to hydrazine or a hydrazine hydrate (hydra JINHIDO lard) (refer to JP,52-22878,B); Carbonic dihydrazide, Screw semicarbazide; A polyfunctional semicarbazide produced by making a hydrazine compound and dihydrazide of the above-mentioned illustration react to a polyisocyanate compound derived from dilsocyanate,

misters (refer to 18,3-18,1358 As and a his parties of this polytunctional semicachaside and a drainage estem hydrophilic radicals, such as polyether polyol and polyethylene-glycols monoalkyl ether, superfluously, Or a isocyanate group in a reactant of this polytsocyanate compound and an active hydrogen compound containing polyfunctional semicarbazide produced by making dihydrazide of the above-mentioned illustration react to an such as hexamethylene di-isocyanate and isophorone diisocyanate, and it superfluously, A drainage system

[0.054]This hydrazine derivative is blended so that 0.01-2 mol of -MH-HM- groups in a hydrazine derivative may be polytunctional semicarbazide, etc. are mentioned.

antiseptic, an antifungal agent, a pH adjuster, a rust-proofer, and a curing catalyst, can be chosen suitably, can be a wetting agent, a defoaming agent, a plasticizer, a film formation auxiliary agent, an organic solvent, an [0055]Further, if needed, additive agents for paints, such as paints, a bulking agent, aggregate, a pigment agent, 0.05-1.5 mol preferably to 1 mol of a carbonyl group contained in said copolymer solution or a water dispersion.

[9900] combined, and can be blended with a distemper constituent of this invention.

[Example]Hereafter, an example is given and this invention is explained still in detail. A "weight section" and "% of

Subsequently, the following monomeric mixture was dropped for 4 hours, maintaining temperature at flowing-back dispersion, and temperature up was carried out, agitating to flowing-back temperature (about 84 **). [0057]Isopropyl 3 stohol 79 weight section was taught into the manufacture example 1 flask of a copolymer water the weight" are meant a "part" and "%", respectively.

temperature.

colonless, almost translucent water dispersion of 10% of the nonvolatile matter was obtained. out temperature up to 50 ** and performing chuming for 2 hours, keeping temperature at 50 **, it cooled, and the dilution tub, after 820 copies of deionized water having added 9.8 copies of triethylamines continuously, carrying solution of 56% of the nonvolatile matter was obtained. Then, the obtained copolymer solution was moved to the temperature after that [one copy], it cools to a room temperature, The almost water-white consistency copolymer Silaplane FM-0711 (notes 2) Ten copies Azobisisobutyronitrile After riping for 2 hours, maintaining at flowing-back Styrene Ten copies 58 copies of n-butyl methacrylate Acrylic acid Seven copies KBM-502 (notes 1) 15 copies [8500]

monomeric mixture, it carried out like Example 1 and the water dispersion of the light opalescence of 10% of a weight 1,000 example 2 Example 1, Except having used the following monoment mixture as a dropped FM-0711 : (Note 1) In the Chisso Corp. make, a poly dimethylsiloxane group content monomer, and molecular [0059]KBM-502:Shin-Etsu Chemical Co., Ltd. make, an alkoxy-silyl-groups content monomer, Silaplane (notes 2)

nonvolatile matter was obtained.

dispersion of the light opalescence of 10% of a nonvolatile matter was obtained. following monomeric mixture as a dropped monomeric mixture, it carried out like Example 1 and the water Silaplane FM-0711 40 copies Azobisisobutyronitrile in one-copy example 3 Example 1, Except having used the Styrene Ten copies 28 copies of n-butyl methacrylate Acrylic acid Seven copies Vinyltrimetoxysilane 15-copy [0900]

Example 1 and the water dispersion of the light opalescence of 10% of a nonvolatile matter was obtained. 1, Except having used the following monomenc mixture as a dropped monomenc mixture, it carried out like Corp. make, a poly dimethylsiloxane group content monomer, and molecular weight 10,000 example 4 Example Chemical Co., Ltd. make, J An alkoxy-silyl-groups content monomer, Silaplane (notes 4) FM-0725 : In the Chisso Silaplane FM-0725 (notes ₄) Ten copies Azobisisobutyronitrile One-copy (notes 3) KBM-503 : [Shin-Etsu Styrene Ten copies N-butyl methacrylate 58 copies Acrylic acid Seven copies KBM-503 (notes 3) 15 copies [1900]

having added 9.8 copies of triethylamines continuously after that and carrying out temperature up to 50 **, and cooled, after performing chuming for 2 hours, keeping temperature at 50 ** after 820 copies of deionized water 93" (Sumitomo 3M make, fluorochemical surfactant) 0.15 copy was added, and it agitated for 10 minutes. It nonvolatile matter was obtained. Then, the obtained resin solution was moved to the dilution tub, the "Fluorad FCmixture as example 6 Example 1, and the almost water-white consistency copolymer solution of 56% of the [0063]The polymerization reaction was performed in the flask by the same operation using the same monomeric translucent water dispersion of 10% of the nonvolatile matter was obtained. triethylamines continuously after that and carrying out temperature up to 50 **, and the colorless, almost chuming for 2 hours, keeping temperature at 50 ** after 820 copies of deionized water having added 9.8 copies of series surface-active agent) 0.4 copy was added, and it agitated for 10 minutes. It cooled, after performing obtained resin solution was moved to the dilution tub, the "KF-355" (Shin-Etsu Chemical Co., Ltd. make, silicone simost water-white consistency copolymer solution of 56% of the nonvolatile matter was obtained. Then, the perfluoreality methacrylate and 60.7 % of the weight of fluorine concentration example 5 Example 1, and the polymentation reaction was performed in the flask by the same operation using the same monomeric mixture as (notes 5) Ten copies Azobisisobutyronitrile One-copy (notes 5) FAMAC : [Nippon Mektron, Ltd. make,] The Styrene Ten copies M-butyl methacrylate 62 copies Acrylic acid Seven copies KBM-502 Ten copies FAMAC [2900]

dropped for 4 hours, maintaining temperature at flowing-back temperature. out, agitating to flowing-back temperature (about 84 **). Subsequently, the following monomeric mixture was [0064]|sopropyl alcohol 79 weight section was taught into the example 7 flask, and temperature up was camed the colodess, almost translucent water dispersion of 10% of the nonvolatile matter was obtained.

natter was obtained. copolymer solution (A), and the almost water-white consistency copolymer solution (B) of 56% of the nonvolatile used the following mixture as a dropped monomeric mixture, the polymerization reaction was performed like the consistency copolymer solution (A) of 56% of the nonvolatile matter was obtained. In another flask, except having flowing-back temperature after that [one copy], to a room temperature. It cooled and the almost water-white KBM-502 Ten copies Silaplane FM-0711 35 copies Azobisisobutyronitrile After riping for 2 hours, maintaining at Styrene Ten copies 30 copies of n-butyl methacrylate Methyl methacrylate Ten copies Acrylic acid Five copies [900]

continuously and carrying out temperature up to 50 **, and the water dispersion of the opalescence of 10% of a temperature at 50 ** after 816.5 copies of deionized water having added 13.3 copies of triethylamines tub, respectively and agitating them for 15 minutes, It cooled, after performing chuming for 2 hours, keeping (A) obtained by the one-copy above like, And after teaching 162 copies of copolymer solutions (B) to a dilution Seven copies Acrylic acid Ten copies KBM-502 15 copies Azobisisobutyronitrile 18 copies of copolymer solutions Styrene Five copies M-butyl methacrylate 26-copy methyl methacrylate 37 copies Acrylic acid 2-hydroxyethyl [9900]

mixture as example 8 Example 1, and the almost water-white consistency copolymer solution (C) of 56% of the [0067]The polymerization reaction was performed in the flask by the same operation using the same monomeric nonvolatile matter was obtained.

copolymer solution (D) of 56% of the nonvolatile matter was obtained. polymerization reaction was performed like the copolymer solution (C), and the almost water-white consistency [0068]In another flask, except having used the following mixture as a dropped monomenc mixture, the nonvolatile matter was obtained.

N-butyl methacrylate 22 copies Methyl methacrylate 48 copies Acrylic acid 30 copies Azobisisobutyronitrile 108 [6900]

triethylamines continuously and carrying out temperature up to 50 **, and the almost water-white water dispersion for 2 hours, keeping temperature at 50 ** after 807.1 copies of deionized water having added 22.7 copies of solutions (D) to a dilution tub, respectively and agriating them for 15 minutes, It cooled, after performing chuming copies of copolymer solutions (C) obtained by the one-copy above like, And after teaching 72 copies of copolymer

deionized water to 10% of the nonvolatile matter, and was used for system performance testing. of the opalescence of 39% of a nonvolatile matter was obtained. The obtained emulsion was diluted with water dispersion was dropped into the flask over 5 hours, it was made to ripe for further 2 hours, and the emulsion of water and distributing the following monomeric mixture, 1.0 copy of ammonium persulfate was added. The the example 9 flack, and temperature up was carried out to 85 **. Then, after using the homomixer for 80 copies [0770]0.15 copy of dodecylbenzenesulfonic acid ammonium and 95 copies of deionized water were taught into of 10% of the nonvolatile matter was obtained.

profitable, generation of a coarse particle and sedimentation took place and the water dispersion of the good state monomeric mixture as a dropped monomeric mixture and the water dispersion was tried with the method of Comparative Example 1 Example 1, Although it carried out like Example 1 except having used the following Acid 1 Part Silaplane FM-0711 10 Part KBM-503 10 Part Dodecylbenzenesulfonic Acid Ammonium In 8.7-Copy n-butyl methacrylate 25.5 copies Acrylic acid n-butyl . Copies [50.5] Acrylic Acid 2-Hydroxyethyl 3 Part Acrylic [1700]

Azobisisobutyronitrile In one-copy comparative example 2 Example 1, Although it carried out like Example 1 Styrene Ten copies 73 copies of n-butyl methacrylate Acrylic acid Seven copies Silaplane FM-0725 Fen copies [0072]

was tried with the method of profitable, generation of a coarse particle and sedimentation took place and the except having used the following monomeric mixture as a dropped monomeric mixture and the water dispersion

water dispersion of the good state was not obtained.

the same operation as Example 1, and the almost water-white consistency copolymer solution of 56% of the monomeric mixture as a dropped monomeric mixture, the polymerization reaction was performed in the flask by Azobisisobutyronitrile in one-copy comparative example 3 Example 1, Except having used the following Styrene Ten copies 73 copies of n-butyl methacrylate Acrylic acid Seven copies Ten copies of FAMAC(s) [6073]

water, and 42 copies of triethylamines and carrying out temperature up to 50 **, and the almost water-white cooled, after performing chuming for 2 hours, keeping temperature at 50 ** after adding 787.8 copies of deionized copies Azobisisobutyronitrile. The resin solution obtained continuously one copy is moved to a dilution tub, It M-butyl methacrylate 22 copies Methyl methacrylate 38 copies Acrylic acid 30 copies Silaplane FM-0711 Ten [4700]

monomeric mixture, it carried out like Example 1 and the water dispersion with almost translucent colonessness [0075]In comparative example 4 Example 1, except having used the following monomeric mixture as a dropped solution of 10% of the nonvolatile matter was obtained.

of 10% of the nonvolatile matter was obtained.

nonvolatile matter was obtained.

.benietdo fon eew

monomeric mixture was dropped for 4 hours, keeping temperature at 95 **. copy comparative example 5 flack, Temperature up was carried out agitating to 95 **. Subsequently, the following KBM-502 15 copies Azobisisobutyronitnie Ethylene-glycol-monobutyl-ether 70 weight section is taught into one-Styrene Ten copies M-butyl methacrylate 48 copies Methyl methacrylate 20 copies Acrylic acid Seven copies [9700]

[4400]

matter was obtained. The obtained resin solution was diluted with deionized water to 10% of the nonvolatile ethylene glycol monobutyl ether, and the almost water-white consistency resin solution of 43% of the nonvolatile to a room temperature, After adding 21 copies of triethylamines as a neutralizer, it diluted with 45 copies of 30 Part Azobisisobutyronitrile After riping for 2 hours, keeping temperature at 95 ** after that [0.7 copy], it cools Methy methacrylate 42.5 copies M-butyl methacrylate 12.5 copies Acrylic acid 4-hydroxybutyl

with delonized water to 10% of the nonvolatile matter, and was used for system performance testing. the emulsion of the opalescence of 39% of a nonvolatile matter was obtained. The obtained emulsion was diluted added. The water dispersion was dropped into the flask over 5 hours, it was made to ripe for further 2 hours, and for 80 copies of water and distributing the following monomeric mixture, 1.0 copy of ammonium persulfate was the comparative example 6 flask, and temperature up was carried out to 85 **. Then, after using the homomixer [0078]0.15 copy of dodecylbenzenesulfonic acid ammonium and 95 copies of deionized water were taught into matter, and was used for system performance testing.

Examples 1-9 and Comparative Examples 3-6 Obtained as 8.7-Copy System-Performance-Testing above, All Acid 1 Part Silaplane FM-0711 20 Part Dodecylbenzenesulfonic Acid Ammonium Water Dispersion or Solution of n-butyl methacrylate 25.5 copies Acrylic acid n-butyl . Copies [50.5] Acrylic Acid 2-Hydroxyethyl. 3 Part Acrylic

** for 2 hours, the following system performance testing was presented. These were painted by a 50-micrometer applicator to the glass plate, respectively, and after making it dry at 100 show good water dispersibility.

[0800](*1) Paint film appearance : viewing estimated and O and a gloss ****** private seal **** thing were made I sldsT ni nwode ei Ilueen A.

gurn meter DCCA type. It is shown that water repellence is so good that the numerical value of an angle of the angle of contact of waterdrop was measured with the harmony chemicals company make KONTAKU tongue [084](*2) Water repellence : the waterdrop of 0.03 cc of deionized water was formed on each coated plate, and into x for the good thing.

[082](*3) Water resisting property : after ****(ing) each coated plate to 20 ** waterworks for 6 hours, what O, a contact is large.

white blush mark, and bilstering are accepted to in what does not have abnormalities in the painted surface was

made into x.

[fable 1] [E800]

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dropped monomeric mixture, it carried out like Example 1 and the colonless, almost translucent water dispersion [0084]]in creation example 10 Example 1 of distemper, except having used the following monomeric mixture as a

[800] of 10% of the nonvolatile matter was obtained.

KBM-502 15 copies Silaplane FM-0711 Ten copies Azobisisobutyronitrile To the water dispersion obtained one Styrene Ten copies 48 copies of n-butyl methacrylate Diacetone acrylamide Ten copies Acrylic acid Seven copies

copy. 5.2 copies of adipic acid dihydrazide was added, it mixed, and the clear coating material was obtained. After painting this by a 50-micrometer applicator to the glass plate and making it dry at 100 ** for 2 hours, when the same system performance testing as the above was presented, paint film appearance and the water resisting property of all were O, and the water contact angle was 93 degrees.

[Function and Effect(s) of the Invention] In the copolymer solution or the water dispersion of this invention. The difficulty of aqueous-izing by the water-repellent group introduced by the monomer (a), and moisture powder, The slkoxy silyl groups introduced into the copolymer by using a monomer (b) as a copolymerization ingredient hydrolyzes under existence of water, and turns into a silanol group, it can conquer, in order that this silanol group may raise water solubility or water dispersibility remarkably, and it can become possible to leasen the amount of copolymerization of the monomer (c) which has a functional group which moreover gives water solution or a dispersibility, and water repellence can be raised. In the drying process of the coat by this copolymer solution or a groups, the coat which shows a good water resisting property is obtained.

[0087] Therefore, the distemper constituent using the copolymer solution or the water dispersion of this invention as a vehicle component can form the coat excellent in water repellence and a water resisting property.

[.enob notalsnerT]

JP11124419 AQUEOUS SOLUTION OR AQUEOUS DISPERSION OF COPOLYME.

COMPOSITION CONTAINING THE SAME WATER-REPELLING PROPERTY, ITS PRODUCTION AND AQUEOUS COATING AQUEOUS SOLUTION OR AQUEOUS DISPERSION OF COPOLYMER HAVING

Publication number IP11124419

11-50-6661

[UAGDJOL: Publication date:

MIYATA NAOKI, NUMA NOBUSHIGE

Applicant:

KANSAI PAINT CO LTD

Classification:

C00D122/00; C08K2/00; C08K250/00; C08K20/00; C08F23/00; C08F22/00; C03D4/00; C00D2/00; C08K2/24' C08E550/10' C08E530/00' C08E33/00' C08T22/00' C03D4/00' C03D2/00' C03D23/00'

- international:

C08E550/45; C08E530/08 C00Dx100; C00D2100; C00D133100; C00D132100; C08E515108; C08E550110; C08E550100; C08E550155; CobD133/06; CobD132/00; (IbC1-1): C08E530/06; C08E550/10; C08E2/54; C08F33/06; C08F32/00; - emobesu:

Priority number(s): JP19970290018 19971022 Application number: JP19970290018 19971022

View IMPADOC patent family

Abstract of JP11124419

eromonom botsunsenu vinolydio- stodo, shqiso tohto lo %.tw 26-0 (A) bas quotg onims bas quotg W. We as despite to measurated monomer having a functional group selected from carboxyl group, sulfonic soid solution is produced by copolymenzing a monomer mixture containing (A) 3-70 Wt.% of one or more more methyl; R \sim 5 is phenyl or a 1-6C alkyl; n<1>1 is 1-10; n<>1 is 5-200) and the formula 1 (R \sim 5 is H or methyl; R \sim 5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or methyl; X is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or methyl); X is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or methyl); X is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or methyl); X is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or methyl); X is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group; (C) 1-25 (R \sim 5 is H or F; n<5 is 1-30), (B) 1-40 Wt.% of a monomer having an alkoxysilyl group. structure, a monomer having an alkexysily! group, an ethylenic unsaturated monomer, etc. SOLUTION: The objective aqueous having excellent water-repellence and water resistance by copolymerizing a mixture containing a monomer having a specific PROBLEM TO BE SOLVED: To obtain the subject aqueous solution useful as a coating resin capable of forming a coating film

丹器開公園出刊時(II)

(A) 舞公指 耕 開 公(21)

(91) 京福舒固本日(61)

特開平11-124419

日11月 2 (6991) 李11 海平 日開公(54)

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数单 图域 套镀链(ZL)		
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强直 田宮 春時発(27)		
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経慮對水む合きけこひ刃、おれ武縄の子ひ刃、強潜化水却>しき蒸衛水枠合血共るできる並水艦 【海各の開発】(43)

樹級購

[181] **`%署藁0∠~ξ**

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[452]

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[185]

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【田跡の來館指計】

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」 J が森木本合重共る 計フい用き玉合重小児多 旅婚代木 よ1>」3 旅客水本台重共の煉場「取求糖」【8 配水精】

を本量単の暦1332な少る化制盤され本量単るれき示 ゔ(11) V及(1) 左査虧の属不(s) 【8000年間】 。去式武場の新増代本より>

これなえ言むる去れ、し、もなるで面の却本楠、 でなる要 金田の場できせる方法もあるが、極端に高い樹脂酸価が必 カウル・カウルはおいてます示き野水焼を分十、下 許多るさサ用動き廃針お面界い高の針木縣、LiT式式の こられなしれる。それでれる示開が去れてかかかるよこ a、8-5とを企業小児多本量単成的不計/シイキエー8、p **メーケしチロクケンサキロシリホしたハた、より了辞公母** さてかりさ エーム平開許払え阀、ブリム母手るで光翔を 点題問ならよのこ。(1) 鎌み槍伏水、小沓木、>蛇は針 木瓶コ級一も間随野木競の来気、れるいろれる68束〉競 ない野水の調陶野水無ならよのこよらな主災胡びま、ら ・位面の主衛の部装並びよは上胡菜汚浸大【EOOO】

むつ消でなるころ科多類並打水班(√高の力熱符水紙)で よいくこらくそ去式るも人等を基型水無い中間断いらよ

の制御小リペイが変く一にいくブリ対いがあった。ふったが

(I) 先查數心語不(B) 、北胛賽本さ間【2000】 。かし、政権の対象にとを見出し本務明に到達した。 ぐ」早付き對水桶で及對水無され勢い鄭道、(1よいくこ) るする取込さ本量単るすする基小しいくくキにハアン及本 量単るや早付き掛木の、果詰なし信納意縫>バを充縛き 殿間に上、よる者門発本【母手の心式るを宏報多題點】 [0000]

後の単量体を3~70重量%、

[9000]

[671]

合重共の毎唱る取水館るもつのよるすする基型水祭るれ (、も示か夫多遊鳌の002~24) 「n、多基 ハキハてのる~「魔案効む又基ハニュてむり、月、中た)

。去て武學の死婚代本はラコュ死部本科

[486]

[83] (。专示 4 天 全 表。ni l41~10の整数で、ni l45~200の衰数 基小キハアのる~1、機累炭は1又基小ニュて了でな異な

$$c_{R}^{s} = c_{C} - c_{C} + c_{R}^{s} + c_{C}^{s} + c_{R}^{s}$$
(II)
(III)

(, 专示 x 夫多機盤の0 E ~[お1 +n ,多茂雲の8~[お1en ,多千原素でてお又 干剤素水よりX 、多基ハキ×よりX干剤素水より 8A 、中洗)

ゴゴを死婚代本はトンプが落水本合産共るでする野水競 るなア」合意共多附合品本量単るを存合%最重己 9~0 チ(b) ひよさ、※量重22~134量単成強不對くし キエー8、20名で育る基準音の断1るれば置る合称の基 、ミヤび近基盤くホ小ス、基小ぐキホ小な(□)、※量 重0 ♪~1を対量単るで育を基小いぐぐキに小で(d)

旗19日本龍るで青舎%量重05~13本量単るでする 基小二ホハカ(9)、竹桝合野州量単 【01原本籠】 。財漁財料並對木るな了し青台フリム代加小の

。附加路序並對水の廣語01頁來需る专声含含 本義稿へいミオゴるす言を基言含sHN-HN-CMS よろな心は中午代1、ブリム降離栗 【11頁水籠】 ,然为路径垄型水(0

アン 3部箇用は並る得し 海ボ多糖並立れ最い針水師で及 野水飛む>し精、J関コ新境代水む>」3 新路水和合風 共る专言多對水船、北阳赛本【理价游技る专屬の肥務】 [[000]

、るす例に特別は特別が代金されてひる。去 式<u>影響の予切及旅</u>盤会水却>Jも<u>新</u>密水本合<u>重共</u>な用す

[0000]

【限號な醂籍の限発】

[4870] [8000] (。中示7天多遊甕000 て~とわ 5 1、1ま1~10の整数を、n2 は5~2 の3~1 凌素気は又基小ニュてファウ異は又一同む 5月 、多基八千×41又干쾲素水約 1.8、中法)【7000】

、多基小卡×LIX干剤紫水L1 EA .中左) 【6000】

$$SH^{2} = C - C - CCH^{2} + CE^{2} + CE^{2} + CE^{2}$$
(II)

J-F8FM, [KZJ-F17FM] (NITALX 現市、3なイーリリクス(タX)ハキエ(ハジテロたハ CAT+1)-5, 1-1119 (0x) NAI(NA 6KOKN(4F642M) -2 , 1-1(197 (8K) 120 COKNCE1 F-8, 8, 2, 2, 1-100 て(をx) 小キエロドハてリイー2,2,2割5周,7 しょことなる。 は単単雄 (14と)の重要での るなる夏不對婚代本計入しる對裔本心なるを夏不心對本 殊六ま、代表が對合重共や社合国の3本量単の例、3.6 太太多05社 in 。各大了城盛の05~11 in ,城盛 へ8~1 計 €n 。るあつ予煎素 v ておけま予刷素水計X 2)において、R3(4本素原子またはメチル基であり、

小てるパち示了(III) 先置都張丁 、お(d) 本量单る でする基へいぐくキにハてブいおこ。開発本【E I O O 】 。るれらや挙むとなどが挙げられる。

、であつのよるも有き基小いぐぐキロ

こじたのとき、R5 同志及びR6 は同志は同じてあって th dn , sha , 专示 x 夫多遊遊の P~ I til dn , 多益小 ジキに小ての01~1. 機案地はX基小キ小ての3~1. 機 素炭 、基小ニュてアック異は又一同は 9月ひ及 5月を基 パキハての01~1燈素数1・3、中た)【さ100】

多示フ(V)込及(Ⅵ) 左査幣 55 T 払 フ 」 5 円表 外 の チ (*いずり)いしゃな話り

。るきブかくこる利率を対量単るな

[9100]

[4815]

- 6) 林量単される示了(11) 大武衛張上【2100】 ころ」(いずれもチッソ社製)等が挙げられる。

、るあてのよるも出張を終加 ・財保室費水るです含てしる代表小でゴゴタ系が代本は1> J·3 旅客水湖合重共越70. 及、去古武蝶の子70. 及, 旅端代 水より>ノも新沓木朴合重共るや斉き針水貎るする端科を 3.1.6.す合重共全(計合配本量単るで)青台%量重 2.6~0 多本量単時館不對くマキエー8, pな対応合意共の断の チ(b) ひよは、沢豊重己2~1多対量単成強不對くし キエー8、204百多基請官の爵士るれ制題と4様の基 しミアV及基盤とホバス、基小ビギホ小な(5)、%量 重りた~13枚量単るでする基小リンジキに小て(b) (。市示が天多機器(OOE~11) *n .多 遊聲の8~1 計En 、多千周案、て却又干剤案木却X

パヤいの(2-6) お量単るを青含を深いてるれち示う (II) <u>た</u> 監 構 に 土 ひ 及 (I ー s) 本 量 単 る を 斉 多 譲 く サ キロぐじれるれる示う(1) 左査構造土、(あつのよる を引入が大型大型大型では、より(s) 対量単の軽 [d 3 >な心るれお題らやお量単される示了(11) ひ及(1) [0100]

ンレーンドM-0721」、「サイラブレーンドM-0 モトセ」、「IIVO-MAベーリアモトセ」、より了品 頭市、ブリム門林具の(1-6) 林屋単遠 いなうしま 望了のるなる身不對婚代本も1>」、4 對答本、、 」 虽不休許 水腺の神合連共制合能るも越交00231並、下れら野が 孙木雅な代十點7節末2社 5n . る杏丁烯醛の0 ≥ I ~ 141~100器数 115 145~500、好事しく1415 らか、必ずしも互いに同じものである必要は無い。 ロ A7基小キルマの3~「凌素気むさま基小ニュてお 5月 、(1名)を小そとおけま予原素水も14名、ブいさこ(1 - 6) 本量単るれる示了(1) 大査結結【1100】 . るちプ用動きれ両却又式 1.か

夫玄基業水外境味鹼類加部の面2の3~1 機素域11 8月 、含基小キ×より又子和紫木む「兄、も示る【2100】

休Aさらの対量単の(VI) 定査附語土六ま【2200】 。687不同の職性みとなくそくぐキイエジハキス 4(3ロベジキャイ(トロ(19で(20×)ーケ , どそジジキ イメジルキメルサロとくジャオルトロじゅて(ダメ)ーァ 、べそくぐチイエじイハコロヤぐキャルトロじりて(を

[9700]

[8200] 10027] TASLOLCIA, MAIR

くじキイエリイルニコ 、ソランシキイメリイルニコエは 3分、払びしる対量単の(V) 大武構造土【OEOO】 、各九分利率が出な【6200】

類木、イーリリクT(ダス) 小キングハジチホリカーさ 、イーリレクア(タメ)ハコロでハジギホリカー2、イ ーリリクマ (ダメ) ハチエバジキホリカー2 . 遡りて て、猫ントリア、猫ンロセト、猫ンイログ、麺小じで下 (そべ) おふ例、おうしる対象単るで言を基小シャホハ の木溶性しくは木分散性を向上させるものであり、か 本合産共 、Ы(o) 本量単成館不型ベンキエー8, nる **すする基消官の動しるパ制鑑さや特の基人ミて**の返基類 くホルス、基小シャホハセブいはご即発本【1600】 、それら利率は2なべそ

 (税) 麺~木小た消言~木無 3 村量単床鉛不るを育き基

大) ーケ 、くそくくキイトじイイツロでくチャルトロじ 4T(4K)-T 、VEVVキイエジキKNトロU4T (0024) - BIRING - (12) 3099847 [D200]

ろなくチャルキクトーロ 、くチャハくチハソト、くチャ

れいキハーロ 、いキインハント 、ジャインハーロ 、ジキ

イヤーナコラナはX-コラミ,-i,-n, ジキホロて

V ト 、 ジキホロてーロ 、 ジキイエ 、 ジキイ×115例 、基

打撃なるないでモーロ 、ハニノーロ 、ハキクボーロ 、小

ものの他に、さらにローヘフ・チル、スーエチルへキン プリ示例フノム基小キハTの3~1.機案数3.A5示了。

れる炭素数1~10のアルキル基としては、Ro 及びR よっていることが挙げられる。 R1 で示さ

ジキハソト、小ジキハーロ、ハキベンヤホ、ルキベツソ

ト、Nキンかーロ、ハキてーナアラナよび一つらさ、一

וי ב+ווי ח-שמצוי אישמצוי ח-, ו

キ×もよ例、基小キ小ての外対代も「X競」、よりブノ 3

る。凡。及び凡。で示される就業数1~6のアルキル基 れる判案をとな基としそメヤチへ、、マリモメをとか、マ V+IN+I , XV+X€1+ , XV+Y-E , SXIX

-E 'I '-Z 'I 'XUBDL 'XUFI 'XUFX

ましては、直鎖又は分枝状のアルキレン基、例えば 木小炭味強熱推韻の耐2の3~1.燃素焼るパち示丁でよ

上記構造式 (IV) 及び(V) において、R3 及びR9 に

(. & 本許多秘意公同与監備や夫 , ti en Vix eA , e

用 , 'A 。专示多 [LIX O Ll an , 含基案水小炭床触熱 税割へ副2へる~1機業拠1。4月、中法)【1200】

いぐキにいての状が代む又酸菌、おフ」と基小ぐキに 小ての0 [~ [機業拠るパさ示了 PAUM BA . される

。るれら刊挙法

[1813]

[8100]

, む1A , 中左) 【 「100】

、る考づなくこるい用ブリ州路 た、これらは1種あるいは2種以上を目的に応じて査宜 され挙みとなくマイスハロペーカ 、くエハイハニコ、く 448114x -0, X448, 11111=01144(4 k) 、小そスエ小ニッロ"Cの当な小ニッロでV A 類指 : 蘇小干スエ小ニンのとない二づ麺へのてた、ハニン麺 語、ハニコ類は、ハニコ類くたコロで、ハニコ類相:既 **ルデーエルニョのとないデーエルニヨルニェて、ルデー** エリニコハシキハロクシ ノハテーエハニコハキクヤ ノハ テーエバニソハシキハ・ハテーエバニソハキとーナ、ハ テーエルニタルキケーロ ノルテーエルニタルタロケイト 、ハデーエルニヨハゴロでーの、ハデーエハニゴハキエ : 蘇小テスエ語小リペア (ペメ)の3な、小テスエのろ 類れいそて(そX) メルートリホルテーエリホのンなれ 一にいかくくりつていれをハーにいかくりもエリホ ノハ チアジキロドコート婚小リヘア(をx)、ハコロアジ キロメコーと強小いので(タメ)、小コロでくきロドコ -2額小じんで(々火)、小キエジキロドコー2鎖小じゃ て(セス)、ハキてジキイエ類小じそて(セス)、ハキ てくキィス強小じてて(ダス)、ハキエくキィエ錯小じ **でて(そと)、小キエジキイ×鏡小じでて(そと)、小** ニハホソト類ハリのT(を木)、ハミキハロのご類ハリ クT(QX)、小UTTX類小UCT(QX)、小Uた そ幼れじゅて(やと)、小心干麺れじゅて(やと)、小 キクヤ-n舖11じクT(タ×) 、いぐキハハキエー2類 れじそて(セス) 、れるそへ猶れじそて(セス) 、れキ てーま猫れじぐて(&×) 、れそてーi櫛れじぐて(& ×) 、ハキて一類ハリカマ (&×) 、ハコロケソト類ハ リカヤ(ガス)、小コロヤーロ鰡小リカヤ(ガス)、小 チエ類1/1/97 (4×) 、ハチ×麵1/1/67 (4×) 、おふ阀、おフリム(b) 料量単成盤不對ソイモエー8 、かな鎖で合選共の曲の子ブバは二肥祭本【2600】 。るれる利率社となるトーンリクで(を

無、猶へハに木無、猶べに冬木木無、猶べトンで木無法 れた、九さ打挙やとな標試付れる今のろ(等猶れをC木 (冬×) -2115門、おフコと本量単るを育金基増へホ たま、るれら刊挙がとな婚へホれたれそエジキロじてて ミアルキメシ訂え間、おフコと本量単るで育金基しミア ドリれキエンミアルキエン、イーリじて「(を×)れそエし メ)れそエトミアれキア、ナーノじて「(を×)

。る考了用動性錯點青の等類得、強小リヤT (やく)、麺指小キメリイ、麹いたコロで、頞相、頞キ 、今朔魏無の等、雄くい、魏版、趙副弘太网、おこ1合称 るやす多基へミでや(こ) 対量単、きつ用動が等ムやい 人が強木、ムヤリイナが強木、アニチング、今ンミアの 2211-16ILETNEIS IN-16ILETNE 大で、、マミアルーしをエリイ、、マミアバーしをエジ、、ベ ミアハーしをエしチ 、ソミサハキケリイ、ソミアハキ ては、マミアハキてしま、ベミアハカロセソトリイ、ベ ミアハコロケイトな 、イミアハコロケイトしチ 、イミ ていキエ(いイ 、ベミヤハキエジ 、ベミヤハキエしチ 、ベ ミヤルキメリイ 、ベミヤルキメジ 、ベミヤルキメしま対 太陽、よい合称るで育る基礎でホバス、基小できホバカ 代水は1>」よ小路木、ブいおご出式伊路本【7600】 10重量器が好ましい。

のみろ。 に理影響をおよばす界面活性別を用いることなく本発明の製造方法によれば解水性や耐水性 のの35]本発明の製造方法によれば解水性や耐水性 の35]本発明の製造方法によれば解水性や耐水性 の35]本発明の製造方法によれば解水性や耐水性 の35]本発明の製造方法によれば解水性や耐水性 の35]本発明の製造方法によれば解水性や耐水性 の35]

4日11-124419

ポル夫を成盤の002~241 「n、き基小キ小での3 ~1 爆業規制又基小ニュて計の「用、中法)【2 400】

にいる数変れそれて、ハトヤソーにいる数変れテーエリ 宋社太网、封二代本具、上17JSIK社計計画界各介多示了 (IV) <u>装造</u>熱 場下 (。 を示か夫 多 成 弦 の 0 E ~ I も 1 8 n 、多子阅蒸、CtlX干 阅案本むY 、中次)【 7 A O O 】

量車01社量成落落。いしま壁ならこされで不以2量車 は、樹脂固形分に対して10重量の以下、摂ましくは5 量加添の液掛計面界結。るれら刊挙は3な直鎖でホ小 スパチルでロおいてーパ、副類へホルな小キ小でロドバ とれる界面活性剤としては、具体的には、例えばパーフ 示フ(IIV)先置動場下、るれる利率オンな小トヤソー

戌與、出衣散釋のQ品前、より了即発本式一【8 4 0 0 】 いなとつま匿込の多 **・
すて型心性水極心及性水無心製並るれる勢くるえ触き**※

本量単品値ブバ用を限が得ごれて、ブリ 4本数を本

、ハテスエハキハア、ハデーエハキハア暦各のソンキエ ジキャリホ、今降針お面界ントニアの3な最強ンホハス 利としては、例えば高級アルコールの硫酸塩、アルキル 小界、るるつ鎖でもとこくな行う影室とるい用多降砂筒 たれる乳化塩合法をも提供するものである。レドックス 合重の対容水、サち小児増代を附合配の(b)~(s)

。るれるい用かるな廃設開入で、イン對畜木 まいるあ、ドンキャーハロドゴンメク、ムケニチンで類 読匙、素水小類配孔太例、おフノ 3体材開合重。るれち 用動化公公廃對活面界對次灵る专言多基环婚不對合重化 及、廃姓活面界ントト非のとなれて一エハリアルキハア

蝶の新雄代本はシ」と新茶本本合重共猛土【0600】 。る太丁のよる下拱點多耐知路符差對水心含 フノム代カルペゴン多新遺代水払>J J 新密水林合重共 るれら武壌で薫の話土、よりて即発本ブい水(9400)

本量単るです
多基本にホルホン・ルカン・カナが高い
と き、Licionも私の(b)~(b) 本量単るれるい用いき

ド、ダイアセトンメタクリルアミド、アセトアセトキジ ミアルリセアイナイトを、マトノロセア割え胸、む ブリュ(ョ) 対量単るで育含基化二ホルル【I200】 。るもでかくこるや存合%量重02~ (も)を、必要に応じて1~30重量%、残ましくは5

断「~♪」ハーロキスハミハホ 、イーソリケを×ハキエ

[8131] [5000]

。る考づ用動祉 のよるす音を基對木無るれる示フ (IIV) よいた (IV)

大登勝場「おより」としては、例えば下記報造者を入りの】 。るきでかくこるも小婚代本も

> J よ外数水されてJ 山流 全廃型活面界る 女育多基對水 鉄口密路合品の(B)部階V及(A)本合重共む>Jと 燕蘇(A) 林合重共、7的目もサち上向き野水餅の開味 漁送観遊3時7 1 網におが合化水よ1> J も外路水の(A)

いなとしま型で **あるるものともいなれる野を担本無な代十くる人数多%量** 重さ94合階用剤の(B) 部樹稿 いしま望みのるもご 6よるなる不以次量重08お2つませ、不以次量重26

コ中代引聞網燈橋合のと(A)本合重共、知合應用動の 【0041】上記機能(B)を用いる場合、機能(B) ・るきかなることがアンコ制制

3 (A) 執合重共和収及合重共 。さきアからこるや用動 多本合重共るれる終でよいくころも合重共プリ税監宜商 ら4中の場形は(b)、(d) 本量単7点(c) 本量単 共多构量单の姚の予心及构量单名专科计多数馆代木入掛 密木は人内、>なお風味に持れれるブのもるす合語代十 3 (A) 本合重共 、おフコ 3 (B) 部路線【0 4 0 0 】

。るるでからこるを登襲 ブノ小遊代水は、ことが溶水ブノ山流を廃床中ひよお水

別代可能な概能(B)格務を混合してから、該連合物に 代水よ1>」と小部水いなま含き基性水類、二面部(A) | 本会産共、おフいはコ宏式開発本立ま【9500】

ことから増払、ゲい化を防止できる。 るくし五本コ宝宝社基パーへそく、ブのるれき合語口約 婚代本が限成中るで進歩き熱深し用計らてしく製餓箱代 木成の基小いぐくキに小ておれよコ玄木のこ。るるで玄 するで加添き廃床中ご参の子、J加添き木ごあ客(A)

林合塵共、比玄市いしませる最もサき遺代木はノ」と小 上記共童合体(A) 密液を、増粘やゲル化させずに水溶 、647台階段Mの8を3内以間和01は1>1ま段、内 以間荷42より3条一、社るな異ファより変態於囲葬、枠 条判別、器容改灵、北間詩の丁生頻遠卧るよコ木らか **山流
麻
味
中
、 よ
に
い
は
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具
。
い
し
ま
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か
よ
こ
と
下
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雷
和
展
展** わけるもうき血流水心及呼中いるかるや血油をパチ 、ラ 反応が急速に進行し、増結、ゲル化するおそれがあるの 木中に徐々に窓加することで相転録することも可能であ 多(A) 本合塵共立」所中、へんるで呱添多水、鋭立で計 多味中プリ血液を開味中心かなし料剤を蒸煮(A) 執合 重共訂え例、合数るを遺化水は1>」よ小密水多(A)執

合産共介れる野ブでよい合産小などで揺上【8600】

ハイチス

、料量単有含基小じぐぐキに小て、寒

【0059】(注1) KBM-502: [[賴/李工業社

10%のほぼ無色の半透明な水分的液を得た。 公発戦不、Jはおみかっている特別問初とさかでき思い

9.8 常然を加えて50°Cに昇温したのち、温度を50°C ベミアハキエリイブハ鯖、猫028水ベヤト棚、J科 こ、関係系を新否本合重共介れる最下い語。介引を新密本 合重共公開部の肥盃自無知別の800代発駐下,JII的 了ま配室教力し、海機間和Sられなる場に表数の子

ルレイニロチ アソトスコンア サイラナレーンドM-OJII KBW-20S (窪I) 題れんして 114K-U額11166X

1148

桝合小イーネマミソトリホるパち軽精でよれ子び返イー ネアジソトジの等イーネアジソトジンロホソトダイーネ ていくんじく マチメサチハ・ドシバルカミサスコ デドジ ミューンでは、 (一とのは、) これを (特別の) という (特別の) モドコリオるなアから成成と (メーモドコマシモドコ) **| 株小木くシミ オーナー・ファンまたはと ドラジン木化物 多基小モスエルキルで級迅速ンホルホ、**ドンモドゴモイ 〒舖エイCナー8, 2, 4, 1, Y ℃ € Y J € √ 〒 婚権 **4ーベンゼントリヒドラジド、エチレンジアミンテトラ** ロトリヒドラジド、クエン酸トリヒドラジド、1,2, **リイニ: オマミオコミイテが大まれでミオコリイ , オマ** モメゴンの鎖イベルメロココひさな、オジモメゴン鎖か タヒソトおけま顔ハをてマモ、鮪ハをて; おびそドコジ 趙くホれなど麻酔不計とトレントしずの等すどそれ」と ン酸ジヒドラジド、フマル酸ジヒドラジド、イタコン酸 トリマ; ドンミド ゴンガンホハル熱視調成菌をを育る干 原素域の間81~2の等インでドゴン難いぐハナ、イン ドラジド、こはく酸ジとドラジド、アジピン酸ジとドラ うじぬいをいた、オマモドラジド、グルタル酸ジビ 【0053】 該とドラジン誘導体としては、例えば落離 ジド基が含まれる。

ハハイミナ、基ドシットリンと きたまには、ヒドラジド基、セミカルバー するとドラジン誘導体を含有することができる。 ここで 1分子中に少なくとも2個の-NH-NH2 含有基を有 、ブリミ解翻架、おい合格される人範34科合重共社基小 二ホハた51、上記帙海路は並却水の肥祭本【2200】

りルアミド、ダイアセトンメタクリルアミドが好適であ トンノ等が挙げられる。このうち特にダイアセトンアク ナルキアハニヨ 、ベイナルキエハニヨ 、ベイヤハキメハ ニコおえ阀) マイヤルチハヤハニコるヤ苷含千部素類の

くそくぐキイメ じょくいこさ 2016677 リキと一点緒小りでをと

[0900]

。六科多新潜代木の当白厚いすらの※01代発戦不、い 行了Jコ祭同 J I 附施実もHtU かい用き附合配本量単 の場下プリ 5 首を配本量単るを干断、アルおコ 1 内部実 SIM越来

張らば

882

10级

照し

0

00、1.量千代、本量単斉含蓋くやキロぐれそんぐじホ 、爆我へぐも:IILO-Wd૮-イムEPH(2玉)

J别

10銀 (2至) J2級 照し 28級

据O I

[8500]

。公J 了断間細り多所

合脈神量単ぽ不るかなさ界コ製脈流敷き変脈でいび、六 J監兵られない行き特徴でま(2048件) 東監流数、A 公計3階量重9 アパーにパアパコロ。アソトコ中にたそで 【例翻天

。各で和意るし※量重!

ひ及し暗量重〕パチパチおし※」ひ及し暗〕。& や肥態 い職籍のるき即終本ブや拳き帆敵実、不以【例赦実】 [9900]

・あきアやくこるを合属了かれ合能し用盤宜産を

廃血添用は整の3.な製盤3.更、降離初、廃盤臨Hq、降 ひな初、隋瀬川、陈密舞寺、廃城朝武、廃壁戸、廃弘祚 、廃断型、廃墟化料額、材骨、廃壊疣、将髄、ブンねつ 要公式るち、おい機動服料塗却水の肥発本【ここのり】 ように配合される。

61~24/1、好ましくは0,05/21、5年/1となる この心基。HN-HN-の中本義務へでミドコブリ校ご 小子【の基小二木小たるれま名の中歌婚代本も1>」と歌 弥木朴合薫共写前、よい本彰続くいで、イゴ鉱【P己00】

。それら刊等は 金(脱級4813284) 4858131-

8年開替) 桝合版のメイジハルなミサ諸盲を系木メイジ ハハセミサ諸百老結よい2歳、ドンハハセミサ諸百老茶木 るれる勢ブサち為東山降戯ネインでドイゴンの示例話上コ 基イーネマジントの中間加減のと耐合外業水型話む含多 基型水脈の等酸ハデーエハキハアしチハーロリヤベンキ エリオや豚ハーセリホハデーエリオろ酵合かイーネでジ V N U 状態、メンジへいなミチ鎖官をるれる野ブサを孤灵 こととうとれる物や上記例示のジとドラジドを通測に

を開発 11-124419	
路○▶	1170-M7<-4
· · · · · · · · · · · · · · · · · · ·	リハビスインプチロニトリル
。立 斜 多	- 1
	英語問3
[1900]	の語イブ」3時合革効量単るや不断、ブバは51月脱粧実
trof 🔨 🏌	デフノコ類同 3.1
路口	<14x
路85	リルチゲーロ鎖リリクを×
異と	類小いへて
避S T	KBW-203 (拝3)
(254) 10部	サイラアレーンFM-0725
12 T	リルフェロチアドトスサンア
A M 基	(牙3) KBW-203:虐線仏⇒工薬杆癬、ブルコチ
の話 T フ 、 3 が合 駅 本量単る す 不 新 、 ブ い は コ 1 例 動実	シシリル 基合有単量体、
デフノス教同 J I 例就実よHUJ SVV用 多桃合風 本量単	(注4)サイラブレーンFM-0725:チッソ社製、
。六野多歌雄代本の西白厚いすらの%01代祭戰不,以	0、01量千代、本量単青含基くサキロぐれそとぐいホ
[0062]	00
10000	√√₹X
婦さ9	ハチと一の緒れじてや木
	• • • • • • • • • • • • • • • • • • • •
場と	強化になる
1 0 部	KBW-20S
路O I	PAMAC (注5)
爆[1/01/2047/77
西無 對對例80名於聲不,以許多表現一	(注5) FAMAC:日本メクトロン社製、バーフルオ
部間づれる軒ブい誘、六軒多族密料合建共な開胡の門表	※量重7 .0 ð 勤勵衆ャて 、 ┦ーリリカを×れギれてロ
溶液を希釈櫓に移し、「フロラードFC-93」(住友	・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・
添き階さ1.0(原針舌面界系素でて、螺状ムエーリス	てつ計製の新同プル用き桝合馬本量単の静同と「例動実
、路028水ン木ト網銭の子。 さつ性期間代の171点	西無知訳の% 3 8 代発戦不 、い 計多辺 3 合重 7 中 に 太 そ
盤いてトリエチルアミンタ、8部を加えて50℃に昇温	間協される斜ブい森。六斜含新弥革合重共な開席の肥蚕
したのも温度を50℃に保ちながら2時間機伴を行って	容務を希釈槽に移し、「KF-355」(信題化学工業
代本な即数半の色無別別の%01代終戰不,J陆的心体	1 ブン山流を踏み、O (所姓活面界系ペービリン、火火
散落を得た。	イブい跡、第028水ベヤト施勢のチ。かり地間間に0
て例献実(4000)	12417731781781781781781781781781781781781781781
びかる路量重9 Γ√ーに√Γ√Γ√Γ り重量部を仕込	温度を50℃に保ちなから2時間機样を行ってから冷却
スプライルを表している。 (484°で) ながら原因 スプライン (484°で) ながら (484°で) と	等多死媒化水な肥恕半の鱼無利和0%01代發戰下,J
合野本量単語するかなる界に変配が数を変配でいる。か	
	 【0063】実施例6
・シント である でんしん しょうしゅん フェック・ファック しょうしゅう しょうしゅう しょうしゅう しょうしゅう しょうしゅう しゅうしゅう しゅうしゃ しゅうしゃ しゅうしゅう しゅうしゅう しゅうしゅう しゅうしゅう しゅうしゅう しゅうしゅう しゅうしゃ しゅうしゅう しゅうしゃ しゅうしゅう しゅうしゃ しゅうしゅう しゅうしゅう しゅうしゃ しゃり しゅうしゃ しゃり しゃり しゃり しゃり しゃり しゃり しゃり しゃり しゃり し	
m≠0: (⊆900)	てブ計製の敷岡ブル用多牌合乱本量単の敷同31円施実
25 O E	7.1+x
260 €	11千七-13個11111111111111111111111111111111111
AFO I	11-4×99/11/06×
2年 2 .	強化になる
類の「	KBW-205
器さら	1170-MAX-14674
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ルチてーロ麺小リクタ×

1148 容林合産共共代以コバル目をは合取場下アンスが合取本量

単るヤ子商、ブバはコにたそくIR。六科多(A) 放容林

台產共な開胡の伊西台無知EIの% 8 2 代班戰不,J 陆的

丁多點室鉄六丁塩焼間部Sされなさ和コ東島前屋銭の予

より2

[9900]

品品

。六科多(B)新商林合重共农商部の即函由無對於O

% 8 8 代発戦不、い行きA及及合産プリコ製同 3 (A) 密

。六つ用コ剱延指サブコ発条コ%01代発戦不ブホント 上棚打く E ぐれて エカれる 野、 力格 多く E ぐれて エの 色 哥多那當代本农肥<u>数鱼無利利</u>(20%01代発戰不,J 政部 白尾の% 6 6 代発戦不下サき海際間部 2 3 0 3 、 3 丁高 のも温度を50℃に保ちながら2時間機样を行ってから コ中にスミてブヤルも問語さる新婚代本の子。 さし 血流を トリエチルアミン22、7部を加えて50℃に昇温した 略0、1ムヤニチンで類節配されてし遺化プロ用きーヤ プい霧、路1、708水ン木下湖、されてJ特散間代さ キミチホい暗08水を附合散本量単の話下ブい際、ぶし I、み込む☆夫の野塚命る昭27(U) 死弥林合重共び 監長のつでる8、不込むを確己を水ン木ト組と暗さ1、0 区にように得られた共重合体溶液(C) 108部、及 I恕 30報 強小してて 路84 **パキ×舖パリクを×** 思るる リイチアーの強いしての大 てつ計製の期間フィノ用を附合版本量単の期間と「開脳薬 [6900] 。六朝多孫趙代木の当白坪の201代孫戰不,J成於 無別別の% 8 2 代発射不 バル計多の気合変すづい期間 3 の方温度を50℃に保ちながら2時間遺程を行ってから トリエチルアミン13、3部を加えて50℃に昇温した 合脈対量単るや下断、ブいはコにたそく限【8300】 ブい蘇、暗さ、318水ベトト船、2ペブン料敷間代さ 透明の粘稠な共重合体溶液(C)を得た。 I、不还出☆夫以幣席条を暗S81(8)新密本合選共 **西無別別の%るさ代発酵不、パ汁を加及合重づ中にたそ** UM、路81(A) 那路本合葉共立なら野いてよいほよ I恕 **ルリ・イニロチてソトスコンて** IP級 KBW-205 10路 類れいてて イチエジキロドコー2額小リケア 出出 路して **パチ×翅バリクや×** (01)6 [Þ Þ Z I - I [小間針

[[[00]]

ムやニチンで鎖ンホハスンサンブいぐディコ中にえそく Q限就実【0 7 0 0 】

8例納実【7000】

路7,8 ムウニチンで競火ホルスマサイブルジデド 娱 0 T KBW-203 얦 0 IサイランレーンドM-OVII 娱 観化してて 煜 ε ハチエビキロドコー2觸ハリクア 据5 .0 5 1/4K-n額1/1/6K 25.5器 **ルキモーロ類小リクを**大

。立つななれる新北部館代水の題外な世見、『こ話体劉 : 放上の干が大併、込むれ続くらよ野き新雄代本、い

○場下ブノ 3 標合具体量単 3 すず新 、 ブバさい 1 内部実 比較例1

I 恕 10報 4437V-VFM-0725 跟し 類れいてて 路ミム **ルキヒーロ錯れじりを**X IO級 114x [0072]

。立た小な九る野山新聞代本の原状な役員、八二届小科 ガ、漁业の干酪大群、かかみ漏るぐよ得多新雄代本、い

行了JSI教局 3 「阿赦実tHttlない用き附合取本量単 の話 下フ しょが 合動 本量単る す 不断 、 ブいさい 1 内 就実 比較例2

提T **ルビイニロキてソトスコツて** 10部 **EVMAC** 盤乙 観れいてて 路とし 1/4と-1.類1/1/66× IO級 1148

[6700]

の話下ブン 3 桝合馬本量単るや下断、ブバおこ1 I 侗鷸実

比較例3

[6800]	オース×キのよるれる体質がわれずい 〇キのよ
・ココス×多のするれる内閣はイヤスや	な独身、J耐喘で射目: 隣代鄭道(1*)【0800】
小白、〇冬のよいなの常異コ面整、鋭さし木好間初るコ	、本示コ15多果結。ふし共の過滤譜的の
【0082】(*3) 耐水性:各墜蒸板を20℃の上水	記す、教式する数類問題2つ7001、J姿整×夫ケー
**	を一代でてmuのさが表えたまされて、Cもプリ示
示さくこるよう役員的本紙野い考大が耐険の食嫌数。 な	多 対 が 代 か な 及 る が す
J支属アコピム DOC - ターメルケンタクをソロ盟北学	の3~6周数出で446月1月1日1日1日1日1日1日1日1日1日1日1日1日1日1日1日1日1日1日
外成認多食嫌野の高木、ノ海消多高水の木くたト湖のっ	性能記錄
端7 .8	チンて麺ンホルスンサンブルンティ
20 宏	1170-M77-17574
路	類れてでて
路 6	ハキエンキロドゴーム麺ハリヘア
焙 ∂.0∈	パキアー ロ 細 小 じ も て
25. 5数	小キとーn 鏑小じぐそと
[6400]	ムウニチンで着ンホルスンサンブルシティコ中にようて
。かい用い線底請封了し席条33%01代発酵不了水く木	[0078] 比較例6
と別れてきくれてエカれる得た。得られたエマルマエの色	10%に希別して性能試験に用いた。
自界の%をそれ発酵不了サき短点間割なこれる多、J不高	代発戦不丁木く木ト組る密部間倒される時。立等多語的
い中に入っている問題と多素強化水のう。立つ血液を	部勝分關於O問悉鱼無別E10% E A 代系戰不了 J 探条 了
席り、「ムヤニチンで独談観みゆう」館代うい用多一や	席とカルデーエルキア(チルーにリヤンリキエ、野ガス
キミチホコ階の8水多桝合鉱料量単の語下7い詩。立し	本名は15ンミマハキエリイフJ 3度成中、J は然フま
0.15部と脱イオン水り5部を仕込み、85℃に昇温	長室教式し、放照商品 るがなる界口の 8 8 多数高級の子
選∠ 0	リリイニロチ アソトスコンア
路 0.6	ハヤアシキロメコーを強いいてて
路 21	類小じでて
40、21	パチてーロ嫋小じぐや木
鳴る . S ♪	小牛×鰡小リカや×
[200]	し監算されない行き特別フまつると、そび出き密量車0
まる 時間 海下した。	トルテーエルキアしチルーにじやくソキエジ中にステく
就合民本量単端下されなさA3O。26全東島でい次。5	许教知 2
器!	パリイニロキてソトスコンて
1 2 銀	KBW-20S
海人	類小しゃす
450公	ハキメ類ハリケチメ
場8₺	11キアーn類111でを×
第01	ソンチス
 	を
f 0 + 0 0 1	是一个一个一个一个一个一个一个一个一个一个一个一个一个一个一个一个一个一个一个
[9400]	
**************************************	数本な限 <u>監</u> の無対表に20%01代発射不、JR的されて。
科多新增代本农阳数半00%014度两个,以	計争特別部とるかなる別のでのである。
示フノコ教同 3.1 開載実むれ以立い用 3 赤合脈 本量単	87、8期、トリエチルアミン42部を加えて50℃に
の話下フ」3就合革本量単るを下高、ブルは31月開新実	「木く木ト組、」等は耐邪条を弦容部的される勢ブい競
路工	リリリーロチアソトスコンア
20年の1	1170-M9<-4-774
海のも	嫌いいてて
2000年8月	いそ×締いいてを× .
髪ここ	ハキて一口類小リヘクメ
[] [] []	西無気灯の% 3 6 代系幹不 、い行 3 成 又合重了中 に 入 こ
,六部多那否本合重共文問初的他委	て予予解の類同31個純実もは似かい用る耐合脈和量単
ተ 81 ላ ማረታሊ 11 ላ መድመር ከመርፎብ የአዲስ 10 በ ነውን	Secured for the last proof of the last of such and the first of the PM of the A TSP (34 pp. 54).

【【表】

[6800]

・カノム×きのよるれる体験ももひをべとした。

I碌

×	×	0	×	0	0	0	0	0	0	0	0	0	發水攝
LL	92	٤9	SL	58	88	88	86	66	83	66	eot		(。) 對水磁
х	0	0	0	0	0	0	0	0	0	0	0	0	器外概型
g	g	Þ	3	6	8	L	9	g	ÿ	ε	г	ĩ	
医糖剂 比較利													

舒多亦遺化木な肥盔半の台無SISIの※0I 代発戰不 、い

[5800]

IO報 路eI 躁し JO恕 路87 IO銀

。るれる野仏観堂を示き卦水

情な役員、他立るで海部多些精緻架な固能でよいの页合 **献**与自の士同基小一へそく。よりアいはつ野衛執政の概並 るよコ新増代本おう」と新啓木本合置共揺さた。るもつ かくこるから上向き對木雅(なる錯になくこるす〉な心 **季量合変共の(□) 村量单るや斉き基請百るや早付き**丼 婚代本へ卦容木むへし、多つ現立コペムるサミ土向>し 巻き封婚代本は>しも卦部本や基ハートそくのこ、でな

盟I

ちついくこる下加引き期差され最い計水桶で放力水粧 木よ1>」も旅客木本合置共の肥発本ファ站【7800】

気引の特益到水【4800】

の話すブリ 5世を指令を単単をです断、ブルはコエ関動薬 0.1例刻美

パキてーn鐕ハリワや× 114x **〒フノン製局 3 1 内部実よけむごかい用き酵舎路料量単**

パリイニロチてソトスヨッて サイラブレーンFM-O711 KBW-205 類れいイイ オミエルリセインイチエトを

3. 68245 9 41 再越弥木、C ようつしバヤバは1卦本簡U及騰代製塾 、そことさし共の機能が動物に共したところ、 に50μmアプリケーターで塗装し、100℃で2時間 承スでひきれこ、六哥を拝堂ーケリペブ」合紙アメ献き 得られた木分散液に、アジピン酸ジヒドラジド5、2部

3基パーしてくて 3 第代水瓜です 五本の水が盛んじくく キロハアゴれち入葬コ中本合産共プムニるやる代気合産 共多(d) 対量単、多らし類の強化本、外部水るよい基 計水銀六パさ人募(よこ)(ら) 本量単 、より了新婚代本よ) >」 3 所称木村合重共の肥発本【果校の肥発び及用計】 [9800]

多類のペーシインロ C

550:45) 90:022

80:212 S30:08Z ZZ:0ZZ \\(C08E 550\10 122/00 00/991 30/661 90/881 00/9 00/9 90/7 C03D 90/ħ COOD Ы 号5個機構 (SI) Int. CI.6